

[barcode]

(19) **FEDERAL  
REPUBLIC OF  
GERMANY**  
[emblem]  
  
**GERMAN  
PATENT OFFICE**

(12) **Unpublished Patent Application**  
(10) **DE 43 31 747 A1**  
(21) File No.: P 43 31 747.2  
(22) Date of application: 09/20/93  
(43) Date of publication: 03/23/95

(51) Int. Cl.<sup>6</sup>:  
**C08 L 97/00**  
B29 B 9/14  
C08 L 97/00  
C08 J 3/12  
C08 L 7/00  
C08 L 89/00  
C08 J 3/18  
C08 J 5/04  
C08 J 9/04  
C08 J 2/20

(71) Applicant:  
  
Wünning, Paul, 78244 Gottmadingen,  
Germany

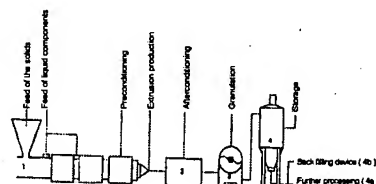
(72) Inventors:  
  
Anonymity requested

Request for examination pursuant to § 44 Patent Act has been filed.

(54) Extruded, fiber-reinforced natural substance granulate for further thermoplastic processing and method for the manufacture thereof.

A natural substance granulate produced by extrusion from predominantly renewable raw materials (lignin, natural rubber, natural fibers, etc.) that may be processed into molded parts like ordinary thermoplastic materials at temperatures of approximately +110°C. Due to the incorporation of inactive pore-forming agents in the natural substance granulate, a pore-forming reaction does not start until the thermoplastic processing, thus making it possible to manufacture low-density molded parts.

The molded parts manufactured from this natural substance granulate are recyclable and/or are naturally decomposable



Legend:

- 1 = Extruder with mechanical devices
- 2 = Climatic zone with mechanical devices
- 3 = Granulator with mechanical devices
- 4 = Storage bin with mechanical devices
- 4a = Further thermoplastic processing with mechanical devices
- 4b = Sack filling/packaging with mechanical devices

[column 1]

#### Description

The present invention relates to a decomposable and compostable natural substance granulate made primarily of renewable raw materials from which it is possible to manufacture molded parts of all types by thermoplastic injection molding methods, as well as the method for its manufacture.

The manufacture of molded parts from natural polymers as described in the patents EP-PS 90 600 and DE 38 27 061/C1 is consistent with the prior art. Similarly (see corporate publication of ICI), it is possible to manufacture molded parts from the plastic granulate offered under the brand name "BIOPOL." Molded parts are also manufactured from plastic granulates based on starch (see corporate publication of NOVON POLYMERS AG/Warner-Lambert Company) and cellulose derivatives (brand name "BIOCETA," for example). All of these known "natural plastics" have more or less the same disadvantages that stand in the way of broad application and accordingly more widespread economic utilization. These disadvantages relate primarily to the high cost of materials (in part 10 to 15 times higher than for synthetic plastics), very high sensitivity to moisture (hygroscopy), which is even increased by amounts of softening agents as well as the in part continued high need for energy (process temperatures are  $\geq 180^{\circ}\text{C}$ ).

The object of the invention is to propose a natural substance granulate that allows low-energy thermoplastic processing in conventional plastics processing machines with controlled reduction of the density and variable moisture resistance without losing the capability of biological decomposability. In addition, in the invention a high value was placed on economy so that the granulate prices are similar to those of the plastics used heretofore. If it is furthermore taken into account that the process temperatures of the natural substance granulate described below are at approximately  $+90^{\circ}\text{C}$  to approximately  $120^{\circ}\text{C}$ , this results in a significant economic advantage due to the lower energy costs. Another economic advantage will derive from the life cycle assessment of the inventive natural substance granulate of the present invention.

According to the invention, the objective is achieved by a natural substance granulate based on polyphenolic adhesives. Such polyphenolic adhesives are produced as recyclings in the production of cellulose. In the thermal conversion process, various proportions of natural proteins (e.g., gelatins), endothermically reacting expansion additives (e.g., foaming agents based on sodium bicarbonate) and softening agents (e.g., water, dihydric or trihydric alcohols) and flexibilizers (e.g., natural rubber) are added to these adhesives. A filler component in the form of natural fibers (e.g., fibers from C4 plants) may be added to the natural substance granulate to increase strength. These fibers are preferably obtained from natural fibers that have exclusively been subjected to mechanical pretreatment. The addition of natural coloring pigments makes coloration possible. The natural substance granulate of the present invention behaves like a synthetic thermoplastic and makes it possible to produce molded objects of all types.

In connection with the examples mentioned and the appended flowchart, the following description of

[column 2]

preferred embodiments of the present invention will further elucidate the invention.

According to the present invention, the natural substance granulate is produced as follows: Starting with commercial recyclings occurring in the production of cellulose fiber, the starting substance is processed, conditioned and granulated into a semi-finished product (strand granulation) according to known methods, e.g., extruders, while adding the additional components cited in the examples such as inactive expansion additives (pore-forming agents), decomposable natural fibers, etc., and further processed in this form as a natural substance granulate into molded parts according to known methods (injection molding machines). In the manufacture of low-density molded parts, it is of the greatest importance that the inactive pore-forming agent is incorporated in the granulate and is present in a uniform distribution. This ensures that the pore-forming reaction does not start before the production of the molded parts and proceeds endothermically.

#### Example 1

The starting material lignin together with a low proportion of gelatin hydrolysate and a proportion of a natural softening agent was fed into the intake opening of an extruder using a mechanical device and melted at a

process temperature of approximately +115°C. At a downstream feeding point, a natural fiber fraction of < 1.6 x 0.4 mm was added to the melt using an additional mechanical device while keeping the process temperature the same. At a further feeding point, the flexibilizer in solution was metered in. After passing through a mixing zone at an equalized temperature, an extrusion was produced via an outlet nozzle, the process temperature having been increased slightly to approximately +125°C. The process temperature had already brought about a corresponding desiccation of the extrusion. After the extrusion was additionally conditioned, it was fed to the granulator using a mechanical device. The mean constant residual moisture of the granulate was approximately 9.5%.

The natural substance granulate thus obtained is very suitable for the thermoplastic manufacture of mechanically loaded molded parts by injection molding.

#### Example 2

The starting material lignin together with a low proportion of gelatin hydrolysate and a natural softening agent component was fed into the intake opening of an extruder using a mechanical device and melted at a process temperature of approximately +110°C. At a further feeding point, the flexibilizer in solution, including the inactive pore-forming agent, was metered in. An extrusion was then produced via an outlet nozzle. The process temperature had already brought about a corresponding desiccation of the extrusion. After the extrusion was additionally conditioned, it was fed to the granulator using a mechanical device. The mean constant residual moisture of the granulate was approximately 10%.

The natural substance granulate thus obtained is very suitable for the manufacture of low-density molded parts by injection molding.

[column 3]

#### Example 3

The starting material lignin together with a low proportion of gelatin hydrolysate and a proportion of a natural softening agent was fed into the intake opening of an extruder using a mechanical device and melted at a process temperature of approximately +115°C. At a further feeding point, the flexibilizer in solution was metered in. After passing through a mixing zone at an equalized temperature, an extrusion was produced via an outlet nozzle, the process temperature having been increased slightly to approximately +125°C. The process temperature had already brought about a corresponding desiccation of the extrusion. After the extrusion was additionally conditioned, it was fed to the granulator using a mechanical device. The mean constant residual moisture of the granulate was approximately 11%.

The natural substance granulate thus obtained is outstandingly suitable for the thermoplastic manufacture of thin-walled molded parts by injection molding.

#### Example 4

The starting material lignin together with a low proportion of gelatin hydrolysate and a proportion of a natural softening agent was fed into the intake opening of an extruder using a mechanical device and melted at a process temperature of approximately +115°C. At a further feeding point, the flexibilizer in solution was metered in. After passing through a mixing zone at an equalized temperature, a lamellar extrusion was produced using a flat nozzle, the process temperature having been increased slightly to approximately +125°C. Using a downstream mechanical device, this extrusion was rolled out further and brought to an appropriate length. The mean constant residual moisture of the granulate was approximately 15%.

The natural substance semi-finished product thus obtained is very suitable for the thermoplastic manufacture of molded parts by hot pressing.

Claims:

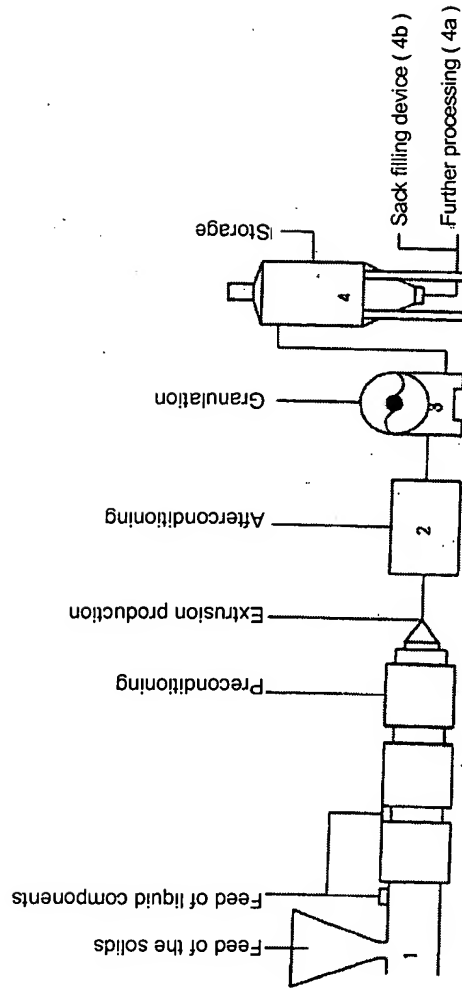
1. Natural substance granulate suitable for thermoplastic processing, characterized in that the starting substance is present as a powdered lignin at a proportion of 25 to 70 percent by weight.
2. Thermoplastic natural substance granulate as recited in Claim 1, characterized in that the starting substance is present as lignin in solution at a proportion of 15 to 70 percent by weight.
3. Thermoplastic natural substance granulate as recited in Claims 1 through 2, characterized by a proportion of 0.1 to 2.5 percent by weight of inactive pore-forming additives.
4. Thermoplastic natural substance granulate as recited in Claims 1 through 2, characterized by a proportion of 1 to 40 percent by weight of natural rubber.
5. Thermoplastic natural substance granulate as recited in Claims 1 through 4, characterized by a proportion of 1 to 20 percent by weight of natural proteins and softening agents.
6. Thermoplastic natural substance granulate as recited in Claims 1 through 5, characterized by a proportion of 0.5 to 50 percent by weight of exclusively mechanically pretreated natural fibers.

[column 4]

7. Thermoplastic natural substance granulate as recited in Claims 1 through 6, characterized by a proportion of 0.05 to 10 percent by weight of natural coloring pigments.
8. Method for manufacturing a natural substance granulate suitable for thermoplastic processing as recited in one of Claims 1 through 7, characterized in that the individual components are fed to the particular mixing zones of an extruder via a feed mechanism and mixed. Via an outlet nozzle of varying geometry, an extrusion is generated that after conditioning is fed to a granulating device and/or granulating system, after which it is to be utilized as a thermoplastic granulate or semi-finished product.
9. Method as recited in Claim 8, characterized in that the process temperatures are between +90°C and +130°C.
10. Method as recited in Claim 8, characterized in that the granulate is conditioned in a climatically constant environment between +25°C and +45°C and a relative humidity between 40% and 65%.

1 page(s) of associated drawings

Annex -1- to the abstract



Legend:

- 1 = Extruder with mechanical devices
- 2 = Climatic zone with mechanical devices
- 3 = Granulator with mechanical devices
- 4 = Storage bin with mechanical devices
- 4a = Further thermoplastic processing with mechanical devices
- 4b = Sack filling/packaging with mechanical devices

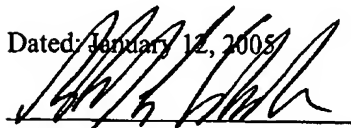


State of New York     )  
                                  )  
County of New York    )     ss:

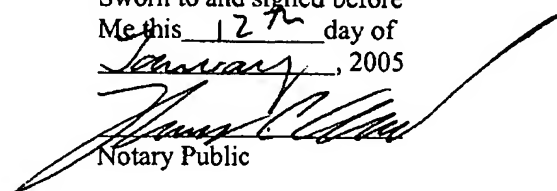
Certificate of Accuracy

This is to certify that the attached document, DE 43 31 747 A1 for Extruded, Fiber-Reinforced Natural Substance Granulate For Further Thermoplastic Processing And Method For The Manufacture Thereof., originally written in German is, to the best of our knowledge and belief, a true, accurate and complete translation into English.

Dated: January 12, 2005

  
Ashley L. Schroeder  
Legal Team Manager, Translations  
Merrill Corporation

Sworn to and signed before  
Me this 12<sup>th</sup> day of  
January, 2005

  
Notary Public

THOMAS C. ALWOOD  
Notary Public, State of New York  
No. 01AL6004438 Qualified in Kings County  
Certificate Filed in New York County  
Commission Expires: 3/26/2008

BEST AVAILABLE COPY